Abstract: In this comment, we argue that the conclusion made by Harrisson and Ben-Yakar [Opt. Express 18, 22556 (2010)], which states that nanoablation with plasmonic nanorods depends on the enhancement of the Poynting vector rather than the one of the square of the electric field, is incorrect and not necessarily needed to explain their experimental results.

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OCIS codes: (350.3390) Laser materials processing; (320.2250) Femtosecond phenomena.

References and links

4. E. Boulais, A. Robitaille, and M. Meunier are preparing a manuscript to be called “Nanorods enhanced femtosecond laser nanoablation,” (2011).

Recently, Harrisson and Ben-Yakar [1] presented a paper on the role of near-field enhancement in laser ablation of a silicon surface using gold nanorods. While very interesting experimental results and calculation are presented, the paper reaches the conclusion that the field enhancement of the Poynting vector rather than the enhancement of the electric field is responsible for laser ablation. We argue in this comment that the use of the Poynting vector is incorrect and in fact not necessary. It can be found in any standard textbook on electromagnetism [3] that, in the case of an harmonic field, the time-averaged rate of work $W$ done on a material contained in a volume $V$, representing the conversion of electromagnetic energy into mechanical or thermal energy, is given by

$$W = \frac{1}{2} Re \left[ \int_V J^* \cdot E \, d^3x \right]$$

(1)

where $J = \sigma E$ is the current distribution in the material. $W$ is obviously proportional to $|E|^2$ when the material is ohmic. Using Maxwell’s equations, basic vector identities and assuming linear and lossless medium, one gets an harmonic version of the Poynting’s theorem which reads

$$Re [\nabla \cdot S] = -\frac{1}{2} Re [J^* \cdot E]$$

(2)
where $S = \frac{1}{2} \mathbf{E} \times \mathbf{H}^*$ is the complex Poynting vector and $\mathbf{E}$ and $\mathbf{H}$ are the complex amplitude of the electric and magnetic field respectively. Equation (2) clearly shows that $|S|$ is not in general a measure of $W$ in the case of an harmonic field. However, $V \cdot S$ would provide the correct result. In the particular case of a plane wave solution propagating in an absorbing medium, $V \cdot S$ is proportional to the Poynting vector itself and $W = \alpha |S|$ where $\alpha$ is the absorption coefficient. The use of $|S|$ to evaluate energy source term for laser ablation is thus only justified in the cases involving plane wave, including for example the interaction of a focused laser on a surface such as in [2]. In the situations involving the interaction with plasmonic nanostructures, the resulting field is by far not a plane wave, as one can easily see by looking at the Mie solution for a sphere in a dielectric medium, and $|S|$ is not a good metric to evaluate energy absorption. This is also true for a plasmonic nanostructure resting on a surface. In [1], both the enhancement of $|S|$ and of $|E|^2$ are evaluated to discuss the power absorption in the nanorod. Two completely different results are found by the authors as seen in Fig. 2. This is not surprising because the fields involved here, especially close to the nanostructure, are not plane waves and the disagreement between the two results only shows that $|S|$ cannot be used in that particular case in evaluating the rate of work done by the field on the material.

The authors in [1] presents two main experimental results supporting the use of the $|S|$ metric. Considering the fact that the use of $|S|$ is theoretically unjustified, we argue that $|E|^2$ can still be the main metric to explain all results. First, they argue that the experimentally measured amplification factor for both nanorod removal ($\sim 89$) and surface ablation threshold ($\sim 92$) is closer to the $|S|$ amplification factor ($\sim 10^2$) than to the $|E|^2$ one. However, Fig. 2 in [1] shows that $|E|^2$ reaches its maximum over an extremely localized area. Laser ablation dynamics is incredibly complex and it is difficult to predict how the material will react to such a localized interaction. Moreover, in the case of the rod ablation itself, majority of the rod rather seems to experience a $|E|^2$ enhancement closer to $10^2$ which would agree much better with the experimental results. It is thus not justified to interpret the maximal field amplification factor directly as an ablation threshold enhancement factor. We therefore think that the use of $|E|^2$ as an ablation metric cannot be ruled out solely based on the arguments presented in the paper. Second, they argue that their failure to observe any double craters shaped hole craters supports the use of the $|S|$ metric. However, recent works in our lab on a very similar system has shown the creation of such a double-hole as shown in Fig. 1. The main difference with the experiment presented in [1] is that the silicon native oxide is removed prior to the nanorod deposition, leading to deeper hole formation. This system however presents a calculated $|E|^2$ very similar to calculation presented in [1]. Shape of the double-hole agrees well with the one of the enhanced $|E|^2$, as shown on Fig. 1. More details will be given in a future publication [4].

Fig. 1. (a) AFM imaging of a double-crater shaped hole created by a single pulse irradiation of a 120 fs, 190mJ/cm$^2$, 800nm laser on a 25nm x 84nm gold nanorod deposited on a silicon surface (sketched in the figure). Laser polarization is linear along the nanorod’s great axis. (b) Cross-section of the double-hole’s depth profile along the line shown in (a).

In conclusion, in addition to being theoretically unjustified, the use of $|S|$ as a metric to evaluate ablation in the case of laser nanoablation with plasmonic nanostructures is not needed to explain their experimental results.